

## (12) UK Patent Application (19) GB (11) 2 253 420 (13) A

(43) Date of A publication 09.09.1992

(21) Application No 9204965.9

(22) Date of filing 06.03.1992

(30) Priority data

(31) 9104781.1 (32) 07.03.1991 (33) GB

(71) Applicant

British Technology Group plc

(Incorporated in the United Kingdom)

101 Newington Causeway, London, SE1 6BU,  
United Kingdom

(72) Inventors

Ian Macmillan Ward

Peter John Hine

Keith Norris

(74) Agent and/or Address for Service

Raymond Hamilton

British Technology Group plc, 101 Newington  
Causeway, London, SE1 6BU, United Kingdom(51) INT CL<sup>5</sup>

D04H 1/54 3/14

(52) UK CL (Edition K)

D1R RFG R308 R309 R310 R314 R551 R553 R562  
B5N N0512 N0526 N175 N176 N178 N179 N18X  
N180 N185 N196 N207 N21Y N223 N225 N227  
N230 N237 N239 N24Y N240 N244 N255 N257  
N259 N26X N2702 N2728 N2730 N2732 N2734  
N2736 N295 N297 N299 N401 N402 N418 N42X  
N42Y N420 N422 N423 N426 N480 N481 N489  
N491 N494 N591 N595 N597 N648 N649 N658  
N66Y N661 N670 N680 N681 N682 N770 N787  
U1S S1034 S1037 S1140

(56) Documents cited

GB 2140837 A GB 1420021 A EP 0354285 A1  
EP 0169184 A WO 88/00258 A1

(58) Field of search

UK CL (Edition K) D1R RFG RFZ RGG RGZ  
INT CL<sup>5</sup> D04H 1/54 1/56 3/14 3/16

(54) Polymeric materials

(57) Polymeric monoliths having high stiffness and strength can be produced by heating an assembly of polymer fibres under a contact pressure to a temperature at which a proportion of the fibre is selectively melted and then compressing the assembly at a higher pressure. Preferably at least 5% of the polymer is melted so that on compression the molten materials fills the voids within the assembly. The use of polyolefin fibres especially melt spun polyethylene fibres is preferred. The products are useful e.g. as orthodontic brackets, bone prostheses and in body armour. The fibres may be in the form of aligned or twisted bundles, chopped fibre, woven mats or layers of fibres at crossing angles.

- 1 -

POLYMERIC MATERIALS

This invention relates to processes for the production of polymer sheet materials from oriented polymer fibres and to the products of such processes.

One method which is widely used to produce high modulus polymer sheets is the formation of fibre reinforced composites using, e.g. oriented polyethylene fibres in order to reinforce the polymer matrix. The manufacture of such composites is a complex operation and in particular requires careful mixing of the polymer and the fibres if the composite is to exhibit homogeneous mechanical properties.

There have been proposals to produce polymeric sheets by compression of networks of polymer fibres at elevated temperatures most notably in relation to thermotropic liquid crystal polymers. European Patent 354285 and United States Patents 4384016 both describe processes in which fibres of a liquid crystal polymer are hot pressed to produce an oriented polymer sheet. European Patent Application 116845, describes a process in which a network of fibres of ultra-high molecular weight polyethylene are hot compressed to form polymer sheets. In the processes taught in this document the fibres are compressed and heated simultaneously. The products retain a significant proportion of the properties of the fibres in the direction in which the fibres are aligned but the mechanical properties of the products in the direction transverse to that in which the fibres are aligned is less than ideal. These processes are relatively unaffected by the choice of compaction temperature. The polymer fibres do not melt during the process.

We have now discovered a novel process whereby an assembly of fibres of oriented polymer may be hot compressed to form a sheet having superior mechanical properties particularly in the direction transverse to that in which the fibres are aligned. The novel processes are distinguished from those of EPA 116845 by an initial processing step in which the fibres are brought to and

held at the compaction temperature whilst subject to a pressure sufficient to maintain the fibres in contact, the contact pressure, and thereafter compacted at a higher pressure, the compaction pressure. In the processes of this invention the  
5 compaction temperature does influence the mechanical properties of the compacted product. In the processes of this invention a proportion of the polymer material in the fibres melts and subsequently recrystallises and it is this melt phase which serves to bind the fibres together.

10 Accordingly from one aspect this invention provides a process for the production of a polymer sheet in which an assembly of oriented polymer fibres is maintained in intimate contact at an elevated temperature sufficient to melt a proportion of the polymer and subsequently compressed so as to produce a coherent  
15 polymer sheet.

In the preferred processes of this invention the conditions and more particularly the temperature at which the fibres are compacted will be such as to cause a portion of the polymer to be selectively melted. On cooling the molten materials recrystallise  
20 to give a phase with a lower melting point than the original fibre. The presence of a second phase in the compacted product may readily be detected e.g. by D.S.C. measurements. In general the amount of material melted is preferably at least 5% and usually at least 10% of the original. The applicants believe  
25 that this minimum amount is required in order fill the spaces between fibres upon compaction and hence produce a product which does not contain trapped air. Processes in which a greater proportion of the polymer material is melted at the contact stage are useful in so far as the mechanical properties of the product  
30 in the direction transverse to the alignment of the fibres may be improved but this improvement is achieved at the expense of the properties in the direction of the alignment of the fibres. We have discovered that the improvements in the transverse direction are not directly proportional to the losses in the direction of  
35 alignment and that the loss is greater than the improvement. For

most applications of the products of this invention the preferred processes are those which are carried out in a manner which selectively melts from 5 to 10% by weight of the polymer material although processes which melt from 10 to 20% by weight of the  
5 polymer or even up to 50% by weight may be useful.

In a preferred embodiment the temperature at which the fibres are compacted is not greater than the peak temperature of melting i.e. the temperature of which the endotherm measured by Differential Scanning Calorimetry (DSC) of the polymer fibres  
10 reaches its highest point. The minimum temperature at which the fibres should be contacted is preferably that at which the leading edge of the endotherm extrapolated to zero intersects the temperature axis.

The pressure at which the assembly of fibres is maintained  
15 during this stage of the process will be such as to maintain the individual fibres in intimate contact but not such as will compact them and in particular not inhibit the selective melting of the polymer. In general pressures in the range 0.5 to 2.0 MPa are preferred. The precise value is not normally critical.

20 The compaction pressure exerted upon the heated assembly of oriented polymer fibres should be sufficient to produce a homogeneous product but should not be so great as to cause the assembly to be extruded. If necessary a closed mould may be used to prevent extrusion and thus allows the use of higher  
25 temperatures or pressures if required. In general, pressures in the range of 40 to 50 MPa have been found to be useful. The minimum pressure required to process an assembly of a particular polymer fibre at a particular temperature may be determined by routine experiment.

30 The time required for the processes of this invention may be determined by empirical means. The time required to bring the assembly of fibres up to the requisite temperature will vary with the nature and size of the assembly, the nature of the polymer and the heating means which are employed. The time is not  
35 critical provided it is sufficient to enable the selective melting to be achieved.

The time required for the compaction step is also non-critical except in so far as it must be sufficiently long to enable the assembly to be compacted. At the preferred temperatures the minimum time may be of the order of seconds  
5 although longer times may be utilised. Processes which utilise shorter compaction times e.g. 5 to 30 seconds may be advantageous in so far as they may conveniently be operated upon a continuous basis for example a uniaxially aligned assembly of heated fibres may be passed between a pair of rollers.

10 The products of the processes of this invention preferably retain at least 50% and more preferably at least 75% of the mechanical properties, especially the modulus of the oriented fibres in the direction in which those fibres are aligned. The products exhibit a homogeneous appearance to the eye. Products  
15 which when stressed in the direction transverse to that in which the fibres are aligned fibrillate, i.e. break whilst leaving the polymer fibres essentially intact are not homogeneous. The products of this invention exhibit homogeneous behaviour when stressed in this transverse direction. Preferably they will be  
20 such that the attenuation of an ultrasonic C scan shows not more than a 20% variation and preferably not more than a 10% variation over the whole sample.

The assembly of oriented polymeric fibres which may be utilised in the processes of this invention may take a variety of  
25 forms. In particular they may be arranged as an uniaxially aligned bundle or a twisted bundle of fibres or an assembly of chopped fibres or as a mat of interwoven bundles or a mat formed by layering of bundles of fibres wherein the bundles in each layer are aligned at an angle, e.g. conveniently perpendicular to  
30 one another. The products obtained by processing such mats may thus retain the majority of the properties of the oriented fibres in more than one direction. The bundles may be assembled and pressed into any convenient shape. The products may be flat sheets, rods, bars, any of which may be shaped so as to be  
35 suitable for particular applications.

The oriented polymer fibres may be obtained by any of the

known manufacturing processes. In particular, fibres which have been produced by melt spinning and drawing and gel spinning and drawing. Typically such fibres will have a diameter in the range 0.005 to 0.05 mm.

5       The processes of this invention may be carried out using conventional equipment. Conveniently, the fibre assembly may be placed in a suitable mould and placed under contact pressure. The assembly may then be preheated to the desired temperature at such a rate as to ensure that there is no significant temperature  
10       gradient across the assembly. The desired compaction pressure is then applied and maintained for sufficiently long for the fibres to cohere. The hot compacted materials are preferably cooled to ambient temperature under controlled conditions. Rapid cooling is less preferred. The most convenient technique is to allow  
15       the compacts to stand in the air until they have cooled to ambient temperature.

      The processes of the present invention may utilise any polymer fibres which can be selectively melted. The susceptibility of particular polymers and particular grades of  
20       that polymer to selective melting varies and their suitability for use in the processes of this invention may be determined empirically.

      The processes of the present invention find particular application in the production of oriented polyolefin articles especially oriented polyethylene articles. The polyethylene (which may be a homo or copolymer of polyethylene) may have a weight average molecular weight  $M_w$  of from 50,000 to 3,000,000. For polyethylene articles the temperature to which the assembly is preheated is preferably within 5°C and more preferably within  
30       2°C of the peak temperature of melting. Oriented polyethylene products of the processes of this invention preferably have a transverse (i.e. in the direction perpendicular to that in which the fibres are aligned) strength of at least 15 MPa and more preferably at least 25 MPa.

35       Gel spun polyethylenes having a weight average molecular

weight of at least 500,000 may exhibit extremely high axial tensile modulus. This corresponds to an extremely high degree of alignment of the polymer molecules within the fibres. These highly oriented gel spun materials may be processed according to this invention and may be preferred where it is desired to produce a product which exhibits high strength in the direction of the fibre alignment. However the strength in the direction transverse to this alignment may be limited unless relatively high proportion of the axial strength is sacrificed by allowing the polymer to melt. Polymer fibres which are not so highly oriented may be preferable in so far as the selective melting which characterises the processes of this invention may affect the axial properties to a lesser degree whilst producing useful strengths in the transverse direction.

Homo and co polymers of polyethylene having a weight average molecular weight of from 50,000 to 500,000 particularly those which can be produced by melt-spinning from a preferred raw material for use in the processes of this invention. Such polymers appear to be more amenable to the selective melting process either by virtue of their comprising some polymer having a relatively low molecular weight or by virtue of their having a surface layer which melts at a lower temperature. Whatever the mechanism which is involved those polymers are preferred because they can form compacts which retain a large proportion of the properties of the fibre (in the direction of alignment of that fibre) whilst producing products having superior properties in the direction transverse to that alignment.

Other classes of polymer fibres which may be useful in the processes of this invention include any of the known orientable polymers. In particular the oriented polymer may be an unsubstituted or mono or poly halo substituted vinyl polymer, an unsubstituted or hydroxy substituted polyester, a polyamide, a polyetherketone or a polyacetal. Suitable examples include vinyl chloride polymers, vinyl fluoride or vinylidene fluoride polymers PHB, PEEK and homo and copolymers of polyoxymethylene.

Particular examples of polyesters useful in the processes of this invention include those derivable by the reaction of at least one polyhydric alcohol, e.g. a linear polyhydric alcohol preferably a diol with at least one poly basic acid, suitably a polycarboxylic acid. The alcohol is preferably an alicyclic or aliphatic alcohol such as cyclohexane-dimethanol or a linear alkylene diol such as ethylene glycol, 1,3 propylene glycol or 1,4 butylene glycol. The preferred acids include o, m or ter phthalic acids, 2,6 and 1,5 napthalene dicarboxylic acid and 1,2 dihydroxy benzoic acid.

The compacted products of the present invention normally have a density less than that of the original fibre. This reduction is caused primarily by the retention of air within the compacted material but also by any reduction in the content of crystalline material within the polymer caused by any molten polymer cooling to form an amorphous phase. Both these factors detract from the properties of the product and the preferred processes of this invention produce products in which the density is at least 90% more preferably at least 95% and most preferably substantially the same as that of the polymer fibre. This reflects the fact that the compaction should preferably be carried out in a manner which expels any trapped air from the product and that in the more preferred embodiment the compact will be cooled in a manner which results in the molten material forming a crystalline phase on cooling.

The processes of this invention enable complicated and precisely shaped polymeric articles having high stiffness and high strength to be manufactured. The products may also exhibit good energy absorbing properties. The products find use in a wide variety of applications, particular examples being as orthodontic brackets, as bone implants and as high impact energy absorbing materials, e.g. in body armour.



The invention is illustrated by the following examples:

The tests used in these examples are defined as follows:

The fibre modulus and strength were measured on a 20cm long sample at a displacement rate of 20 cm/min.

5 The flexure modulus of the samples produced from the process were measured under the guidelines of ASTM D790.

The flexure strengths of the samples produced from the process were measured under the guidelines of ASTM D790.

10 The short beam shear strength of the samples measured under the guidelines of ASTM D2344.

The densities of the compacted materials were measured using a density bottle.

15 Ultrasonic elastic properties were measured using an immersion method at a frequency of 2.25 MHz. A full description of the technique can be found in S.R.A. Dyer, D. Lord, I.J. Hutchinson, I.M. Ward and R.A. Duckett, J. Phys. D:Apply. Phys. 25 (1992) 66.

The fibres used were polyethylene fibres having the following particulars:

	Sample	Fibre	Molecular Weight		Process	Breaking Strength	Tensile modulus	
			Mw	Mn			initial secant	2%
						GPa	GPa	GPa
25	1	CELANESE (R.T.N.)	61,000	28,000	melt spun	1.0	54	36
30	2	SNIA (R.T.N.) FIBRE	130,000	12,000	melt spun	1.3	58	43
	3	TEKMILON (R.T.N.)	700,000	54,000	solvent spun	2.1	80	70
35	4	SPECTRA 1000	1,500,000	75,000	gel spun	2.9	130	115

#### Example 1

40 A sheet of dimensions 3mm x 5cm x 10cm was prepared by hot pressing a unidirectionally aligned bundle of melt spun SNIA (R.T.N.)/high modulus polyethylene fibres having a diameter of 0.015mm in an

open ended matched metal mould. The fibres were preheated for 10 minutes under contact pressure of 0.5MPa at  $139 \pm 0.5^\circ\text{C}$  and then a pressure 400MPa was applied for 10 seconds. The resulting product was a homogeneous translucent sheet with the following properties.

5	Tensile modulus in fibre direction	57 GPa	measured
	Transverse to fibre direction	4.2 GPa	ultrasonically
10	Flexure modulus in fibre direction	35 GPa	ASTM D790
	Transverse to fibre direction	3.2 GPa	
	Short beam shear strength	29 GPa	ASTM D2344
15	Flexure strength in fibre direction	110 MPa	ASTM D790
	Transverse to fibre direction	31 MPa	

An ultrasonic immersion 'C' scan of the product showed only a 2% change in attenuation over the sample and is taken as a measure of the homogeneity of the product.

A DSC trace of the compacted material showed that 8% of the original fibre phase had been melted and had recrystallised forming a second lower melting point phase.

The density of the compacted material was 90% of the original fibre density.

#### Example 2

A bar of 3mm square cross section was prepared by hot pressing a twisted bundle of melt spun SNIA<sup>(R.T.M.)</sup> high modulus polyethylene fibres having a diameter of 0.015mm in an open ended matched metal mould. The fibres were preheated at  $139 \pm 0.5^\circ\text{C}$  for 10 minutes and then pressed for 30 seconds at a pressure of 50 MPa. The resulting product was a homogeneous translucent bar with a flexural modulus (ASTM D790) of 32 GPa.

#### Example 3

An orthotropic material was made by compacting a number of layers of a woven mat of melt spun SNIA<sup>(R.T.M.)</sup> high modulus polyethylene fibres in an open ended matched metal mould. The laminated mat was maintained at  $139 \pm 0.5^\circ\text{C}$  for 10 minutes at 0.5 MPa before applying a high pressure of 50 MPa for 30 seconds. The flexure

modulus was the same in both the axes in the plane of the plate, with a value of 11 GPa. The flexure strength was also similar in the two axes in the plane of the plate with a value of 85 MPa. We can conclude that using a woven mat for compaction results in a substantial improvement in transverse strength at the expense of stiffness.

Example 4

A three dimensional shape was formed by compacting a number of layers of a woven mat of melt spun SNIA<sup>(R.T.M.)</sup> high modulus polyethylene fibre between male and female hemispherical moulds. The compaction conditions were identical to those shown in example 3. The compacted material was formed into the required shape in a single process.

Example 5

A laminated sheet 3mm thick and 55mm square was made by sandwiching a uniaxially aligned bundle of melt spun SNIA<sup>(R.T.M.)</sup> polyethylene fibres between two layers of a woven mat of melt spun SNIA<sup>(R.T.M.)</sup> polyethylene fibres. The sandwich was then compacted using conditions given in example 3. The result was a translucent sheet with the following properties.

	Tensile modulus in fibre direction	52 GPa	measured
	Transverse to main fibre direction	4.9 GPa	ultrasonically
25	Flexure modulus in main fibre direction	18 GPa	ASTM D790
	Flexure strength transverse to main fibre direction	75 GPa	ASTM D790

Lamination allows a better compromise to be achieved between stiffness and strength, especially in tension.

Example 6

2.0 grams of chopped melt spun SNIA<sup>(R.T.M.)</sup> high modulus polyethylene fibre was placed in a cylindrical mould which was 12mm in diameter and 30mm long. Compaction of the fibre assembly proceeded according to the conditions described in example 3. The resulting cylindrical bar was an isotropic material having a modulus of 5 GPa. A DSC trace of the product showed that 12% of the original fibre had been melted.

Example 7

A bar of 25mm square cross section and 100mm long was prepared by hot pressing a number of cold compacted layers of melt spun SNIA<sup>(R.T.M.)</sup> high modulus polyethylene fibres in a closed  
5 matched metal mould using conditions described in example 3. DSC traces taken through the compacted blocks showed that a reasonably even heat distribution had been achieved.

Example 8

3.0 grams of melt spun CELANESE<sup>(R.T.M.)</sup> high modulus polyethylene  
10 fibre with a diameter of 0.015mm was compacted in an open ended rectangular section steel mould at a compaction temperature of  $134 \pm 0.5^\circ\text{C}$ . A contact pressure of 0.5 MPa was held for 10 minutes and then a pressure of 40 MPa was applied for 30  
15 seconds. The sample had the appearance of a solid polyethylene rod with a well defined cross section measuring 3.34mm x 3.11mm. The bending modulus was 19.7 GPa.

Example 9

To demonstrate the criticality of the moulding temperature, a sample identical to that used in example 8 was compressed in the  
20 same mould at the higher temperature of  $138^\circ\text{C}$ . The resulting sample again had the appearance of a solid polyethylene rod but the low bending modulus of 1.2 GPa showed that the properties of the fibre had been lost due to substantial melting of the  
25 the fibre phase. Further evidence of the critical nature of the temperature was shown by compressing an identical sample to examples 8 and 9 but at the lower temperature of  $127^\circ\text{C}$ . The resulting product had a high stiffness but poor transverse properties due to almost total retention of the original fibre phase.

30 Example 10

The role of pressure was examined by carrying out an identical experiment to example 1 except that high pressure (40 MPa) was applied from the very start of the procedure, including the warm up period. The resulting product had a high  
35 longitudinal stiffness of 60 GPa but a poor transverse strength

of 12 MPa. A DSC trace of the compacted material showed no evidence of any 'second phase': the compacted material was composed entirely of the original fibre phase.

5 We can therefore conclude that applying high pressure from the beginning of the compaction process inhibits the selective melting which is necessary for optimum control of the properties of the final product.

Example 11

10 A sheet of dimensions 3mm x 55mm x 55mm was prepared by compacting a unidirectionally aligned bundle of gel spun SPECTRA high modulus polyethylene fibres in a matched metal mould. The processing conditions were identical to example 3 apart from raising the compaction temperature to  $152 \pm 0.5^{\circ}\text{C}$ , which is midway between the onset of melting and the end of melting.

15 The resulting compacted sheet was homogeneous and had a longitudinal modulus of 35 GPa and a transverse strength of 17 MPa. A DSC trace of the compacted material showed around 35% of a 'second phase' formed by melting of the original fibre.

CLAIMS

1. A process for the production of a homogeneous polymeric monolith characterised in that an assembly of oriented polymer fibres is maintained in intimate contact at an elevated  
5 temperature sufficient to melt a proportion of the polymer and subsequently compressed.
2. A process according to Claim 1 characterised in that the assembly is maintained at a temperature at least that at which an extrapolation of the leading edge of the endotherm of the  
10 oriented fibres measured by differential scanning calorimetry intersects the temperature axis.
3. A process according to either of Claims 1 to 2 characterised in that the temperature at which the assembly is maintained is less than the peak temperature of melting of the polymer fibres  
15 as measured by differential scanning calorimetry.
4. A process according to any of Claims 1 to 3 characterised in that the assembly is maintained at a temperature such that at least 5% by weight of the polymer is melted.
5. A process according to any of the preceding claims  
20 characterised in that the assembly is maintained at a temperature such that no more than 20% by weight of the polymer is melted.
6. A process according to either of Claims 4 or 5 characterised in that the assembly is maintained at a temperature such that  
from 5 to 10% by weight of the polymer is melted.
- 25 7. A process according to any of the preceding claims characterised in that the assembly is maintained under contact pressure.
8. A process according to any of the preceding claims characterised in that the heated assembly is compressed under a  
30 compaction pressure of from 0.5 to 2.0 MPa.
9. A process according to Claim 8 characterised in that the compaction pressure is maintained for a period of at least 5 seconds.
10. A process according to any of the preceding claims  
35 characterised in that the hot compressed material is allowed to

cool to ambient temperature by standing in air.

11. A process according to any of the preceding claims characterised in that the polymer fibres are homo or copolymers of a polyolefin.

5 12. A process according to Claim 11 characterised in that the polymer fibres are polyethylene fibres.

13. A process according to either of Claim 11 or 12 characterised in that the polymer has a weight average molecular weight of from 50,000 to 3,000,000.

10 14. A process according to any Claims 11 to 13 characterised in that the polymer has a weight average molecular weight of from 500,000 to 3,000,000.

15 15. A process according to any of Claims 11 to 13 characterised in that the polymer has a weight average molecular weight of from 50,000 to 300,000.

16. A process according to any of Claims 11 to 15 characterised in that the fibre is a melt spun fibre.

20 17. A process according to any of Claims 11 to 16 characterised in that the assembly is maintained at a temperature which is no more than 5°C below the peak temperature of melting of the polymer fibre.

25 18. A process according to any of Claims 11 to 17 characterised in that the assembly is maintained at a temperature which is no more than 2°C below the peak temperature of melting of the polymer.

19. A process according to any of the preceding claims characterised in that the compressed product has a density which is at least 90% of the original fibre density.

30 20. A process according to any of the preceding claims substantially hereinbefore described with reference to the foregoing examples

91MAR

ID95

- 15 -

**Patents Act 1977**  
**Examiner's report to the Comptroller under**  
**Section 17 (The Search Report)**

Application number

9204965.9

**Relevant Technical fields**

(i) UK Cl (Edition K ) D1R (RFG, RFZ, RGG, RGZ)

(ii) Int Cl (Edition 5 ) D04H 1/54, 1/56, 3/14, 3/16

**Databases (see over)**

(i) UK Patent Office

(ii)

Search Examiner

ALEX LITTLEJOHN

Date of Search

8 APRIL 1992

**Documents considered relevant following a search in respect of claims**

1-20

Category (see over)	Identity of document and relevant passages	Relevant to claim(s)
X	GB 2140837 A (ALLEN) See whole document, especially page 5 lines 65-99	1-19
X	GB 1420021 (KIMBERLY-CLARK) See especially page 3 lines 29-36 and 40-50	1, 4-8, 11-16
X	EP 0354285 A1 (DU PONT) See especially examples 6, 7, 8	1-19
X	EP 0169184 A1 (MOLNLYCKE) See especially page 6 line 29 - page 7 line 28	1-6, 8, 10-19
X	WO 88/00258 A1 (BURNETT) See especially page 10 line 11 - page 11 line 7	1-19



Category	Identity of document and relevant passages	Relevant to claim(s)

**Categories of documents**

X: Document indicating lack of novelty or of inventive step.

Y: Document indicating lack of inventive step if combined with one or more other documents of the same category.

A: Document indicating technological background and/or state of the art.

P: Document published on or after the declared priority date but before the filing date of the present application.

E: Patent document published on or after, but with priority date earlier than, the filing date of the present application.

&: Member of the same patent family, corresponding document.

**Databases:** The UK Patent Office database comprises classified collections of GB, EP, WO and US patent specifications as outlined periodically in the Official Journal (Patents). The on-line databases considered for search are also listed periodically in the Official Journal (Patents).

